Observation of dark state polariton collapses and revivals

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By time-dependent variation of a control field, both coherent and single photon states of light are stored in, and retrieved from, a cold atomic gas. The efficiency of retrieval is studied as a function of the storage time in an applied magnetic field. A series of collapses and revivals is observed, in very good agreement with theoretical predictions. The observations are interpreted in terms of the time evolution of the collective excitation of atomic spin wave and light wave, known as the dark-state polariton.

Atomic ensembles show significant promise as quantum memory elements in a quantum network [1, 2, 3, 4, 5]. A "dark-state polariton" is a bosonic-like collective excitation of a signal light field and an atomic spin wave [6], whose relative amplitude is governed by a control laser field. In the context of quantum memories, the dark state polariton should enable adiabatic transfer of single quanta between an atomic ensemble and the light field. Seminal "stopped-light" experiments that used laser light excitation [7, 8, 9] can be understood in terms of the dark-state polariton concept. In a recent work the storage and retrieval of single photons using an atomic ensemble based quantum memory was reported, and the storage time was conjectured to be limited by inhomogeneous broadening in a residual magnetic field [10].

We have recently predicted that dark-state polaritons will undergo collapses and revivals in a uniform dc magnetic field [11]. During storage, the dark-state polariton consists entirely of the collective spin wave excitation. According to the dark-state polariton concept, the retrieved signal field should exhibit the collapse and revivals experienced by the spin wave. The revivals occur at integer multiples of one half the Larmor period, with dynamics that are sensitive to the relative orientation of the magnetic field and the light wavevector. The spin wave part of the dark-state polariton involves a particular superposition of atomic hyperfine coherences (see Eq.(1) below), intimately related to the phenomenon of electromagnetically-induced transparency (EIT) [12, 13]. Revivals of single atom coherences were observed in atom interferometry [14, 15]. Coupled exciton-polariton oscillations in semiconductor microcavities have also been reported [16, 17].

The remarkable protocol of Duan, Lukin, Cirac, and Zoller (DLCZ) provides a measurement-based scheme for the creation of atomic spin excitations [3]. In systems where EIT is operative, these excitations will in general contain a dark-state polariton component. The orthogonal contribution may be regarded as a bright-state polariton in that it couples dissipatively to the excited atomic level in the presence of the control field [18]. Observation of the retrieved signal field, however, picks out the dark state polariton part, while the orthogonal component is converted into spontaneous emission [11]. A number of previous works reported generation and subsequent retrieval of DLCZ collective excitations [19, 20, 21, 22, 23, 24, 25, 26, 27]. Several of these studies investigated the decoherence of these excitations in cold atomic samples [19, 23, 24, 26, 27]. It has been similarly conjectured in these works that the decay of the coherence was due to spin precession in the ambient magnetic field. While the observed decoherence times are consistent with the residual magnetic fields believed to be present, the observation of revivals predicted in Ref.[11] would be solid proof that Larmor precession is indeed the current limitation on the quantum memory lifetime. Moreover, controlled revivals could provide a valuable tool for quantum network architectures that involve collective atomic memories [3, 4, 5].

With this goal in mind, we report in this Letter observations of collapses and revivals of dark-state polaritons in agreement with the theoretical predictions [11]. In our experiment, we employ two different sources for the signal field, a coherent laser output and a conditional source of single photons [3]. The latter is achieved by using a cold atomic cloud of ^{85}Rb at Site A in the off-axis geometry pioneered by Harris and coworkers [25]. Another cold atomic cloud of ^{85}Rb at Site B serves as the atomic quantum memory element, as shown in Fig. 1. Sites A and B are physically located in adjacent laboratories connected by a 100 meter long single-mode optical fiber. The fiber channel directs the signal field to the optically thick atomic ensemble prepared in level $|b\rangle$. The inset in Fig. 1 indicates schematically the structure of the three atomic levels involved, $|a\rangle$, $|b\rangle$ and $|c\rangle$, where $\{|a\rangle; |b\rangle\}$ correspond to the $5S_{1/2}, F_a = 3, F_b = 2$ levels of ^{85}Rb, and $|c\rangle$ represents the $5P_{1/2}, F_c = 3$ level associated with the D_1 line at 795 nm. The signal field is resonant with the $|b\rangle \leftrightarrow |c\rangle$ transition and the control field with the $|a\rangle \leftrightarrow |c\rangle$ transition.

When the signal field enters the atomic ensemble at Site B, its group velocity is strongly modified by the control field. By switching off the control field within about 100 ns, the coupled excitation is converted into a
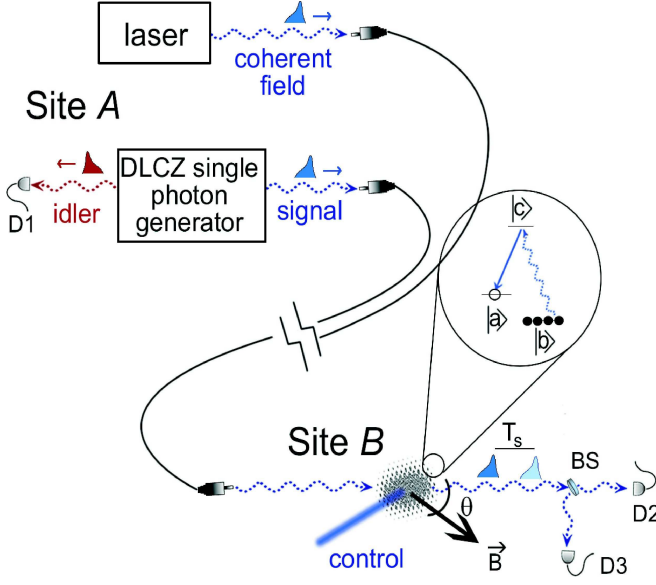
FIG. 1: A schematic diagram illustrates our experimental setup. A signal field from either a laser, or a DLCZ source of conditional single photons at Site A is carried by a single-mode fiber to an atomic ensemble at Site B, where it is resonant on the $|b\rangle \leftrightarrow |c\rangle$ transition. The signal field is stored, for a duration T_s, and subsequently retrieved by time-dependent variation of a control field resonant between levels $|a\rangle$ and $|c\rangle$. All the light fields responsible for trapping and cooling, as well as the quadrupole magnetic field in the MOT, are shut off during the period of the storage and retrieval process. An externally applied magnetic field created by three pairs of Helmholtz coils (not shown) makes an angle θ with the signal field wavevector. The inset shows the structure and the initial populations of atomic levels involved. The signal field is measured by detectors D2 and D3, while detector D1 is used in the conditional preparation of single photon states of the signal field at Site A.

spin wave excitation with a dominant dark state polariton component, i.e., the signal field is "stored" [6, 7, 8, 9]. An important condition to achieve this storage is a sufficiently large optical thickness of the atomic sample, which enables strong spatial compression of the incident signal field [5]. In our experiment the measured optical thickness $d \simeq 8$. The subsequent evolution of a dark state polariton in an external magnetic field is predicted to reveal a series of collapses and revivals whose structure is sensitive to the magnitude and orientation θ of the field relative to the signal wavevector [11].

As we deal with an unpolarized atomic ensemble, we must take into account the Zeeman degeneracy of the atomic levels. Choosing the same circular polarizations for both the probe and the control fields allows us to retain transparency [10]. For a σ_+ polarized signal field, the dark state polariton annihilation operator for wavenum-

ber q is given by [11]

$$\hat{\Psi}(q,t) = \frac{\Omega(t)\hat{a}_{k,+} - \sqrt{Np}g^* \sum_m R_m \hat{S}_{a\ m}^{b\ m}(q,t)}{\sqrt{|\Omega(t)|^2 + Np|g|^2 \sum_m |R_m|^2}} \quad (1)$$

where $\Omega(t)$ is the control field Rabi frequency, g the coupling coefficient for the signal transition, m is the magnetic quantum number, $R_m = C_{m\ 1\ m+1}^{F_b\ 1\ F_c}/C_{m\ 1\ m+1}^{F_a\ 1\ F_c}$ is a ratio of Clebsch-Gordan coefficients, N is the number of atoms, $p = 1/(2F_b+1)$, $\hat{a}_{k,+}$ is the field annihilation operator for the mode of wavevector $k = q+\omega_0/c$ and positive helicity, ω_0 is the Bohr frequency of the $|b\rangle \leftrightarrow |c\rangle$ transition, $S_{a\ m}^{b\ m}(q,t) \equiv 1/\sqrt{Np} \sum_\mu \hat{\sigma}_{b\ m,\ a\ m}^{(\mu)}(t)\exp(-i(qz_\mu - \Delta(t - z_\mu/c)))$ is a collective spin wave operator, where $\hat{\sigma}_{b\ m,\ a\ m}^{(\mu)}(0) = |b,m\rangle_\mu\langle a,m|$ is a hyperfine coherence operator for atom $\mu = 1,..N$, z_μ is the position of atom μ, and Δ is the hyperfine splitting of the ground state. When R_m is finite for all m, the atomic configuration supports EIT, but when one or more R_m is infinite, there is an unconnected lambda configuration, EIT is not possible and dark state polaritons do not exist. Specifically, the excited state $|c,m+1\rangle$ is not coupled by the control field to a state in the ground level $|a\rangle$. An atom in the state $|b,m\rangle$ would absorb the signal field as if no control field were present.

The signal is stored in the form of spin wave excitations associated with the dark state polaritons $\sim \sum_m R_m \hat{S}_{a\ m}^{b\ m}(q)$ for some range of q's. During the storage phase, and in the presence of the magnetic field $\mathbf{B}$, the atomic hyperfine coherences rotate according to the transformation

$$\hat{S}_{am}^{bm}(q,t) = \sum_{m_1=-F_b}^{F_b}\sum_{m_2=-F_a}^{F_a} \mathcal{D}_{m_1 m}^{(b)\dagger}(t)\mathcal{D}_{m,m_2}^{(a)}(t)\hat{S}_{am_2}^{bm_1}(q,0), (2)$$

where $\mathcal{D}_{m,m'}^{(s)}(t) \equiv \langle s,m|\exp(-ig_s\mathbf{\Omega}\cdot\hat{\mathbf{F}}t)|s,m'\rangle$ is the rotation matrix element for hyperfine level s, $\hat{\mathbf{F}}$ is the total angular momentum operator, $\mathbf{\Omega} \equiv \mu_B\mathbf{B}/\hbar$, μ_B is the Bohr magneton, g_a and g_b are the Landé g factors for levels $|a\rangle$ and $|b\rangle$ of ^{85}Rb and, ignoring the nuclear magnetic moment, $g_a = -g_b$. This rotation dynamically changes the dark state polariton population during storage.

The measured signal retrieved after a given storage time T_s is determined by the remaining dark state polariton population. Stated differently, only the linear combination of hyperfine coherences $\sim \sum_m R_m \hat{S}_{a\ m}^{b\ m}(q, T_s)$ contributes to the retrieved signal. We calculate the number of dark state polariton excitations as a function of T_s using Eqs.(1) and (2), $\langle\hat{N}(T_s)\rangle = \sum_q\langle\hat{\Psi}^\dagger(q,T_s)\hat{\Psi}(q,T_s)\rangle$, and find

$$\frac{\langle\hat{N}(T_s)\rangle}{\langle\hat{N}(0)\rangle} = \left|\sum_{m_1 m_2}\frac{R_{m_1}R_{m_2}}{\sum_m |R_m|^2}\mathcal{D}_{m_2 m_1}^{(b)\dagger}(T_s)\mathcal{D}_{m_1 m_2}^{(a)}(T_s)\right|^2. (3)$$

In Fig. 2, panels (f) through (j), we show the retrieval efficiency for various orientations of a magnetic field of

magnitude 0.47 G, corresponding to the Larmor period of 4.6 μs. With the approximation $g_a = -g_b$ it is clear that the system undergoes a revival to the initial state after one Larmor period $(2\pi/|g_b\mathbf{\Omega}|)$, and thus the signal retrieval efficiency equals the zero storage time value. Depending on the orientation of the magnetic field, a partial revival at half the Larmor period is also observed. For a magnetic field oriented along the z axis (Fig. 2(f)), the polariton dynamics is relatively simple. Each hyperfine coherence $\hat{S}^b_{a\ m}^{\ m}$ merely picks up a phase factor that oscillates at frequency $2m|g_b\mathbf{\Omega}|$, thus returning the system to its initial state at half the Larmor period. In this case, the partial revival is actually a full revival. On the other hand, for $\theta = \pi/2$ (Fig. 2(j)), a rotation through half the Larmor period causes the coherence transformation $\hat{S}^b_{a\ m}^{\ m} \rightarrow -\hat{S}^b_{a\ -m}^{\ -m}$, and as a result, the retrieval efficiency is reduced to $(\sum_m R_m R_{-m}/\sum_m R_m^2)^2$. The substructure within a half Larmor period is associated with interference of different hyperfine coherences contributing to the dark-state polariton [11].

To test these predictions, we apply a uniform dc magnetic field of magnitude 0.5 ± 0.05 G to the atomic ensemble using three pairs of Helmholtz coils. In our first set of measurements, 150 ns long coherent laser pulses containing on average $\simeq 5$ photons serve as the signal field. The outputs of the single-photon detectors D2 and D3 are fed into two "Stop" inputs of a time-interval analyzer which records the arrival times with a 2 ns time resolution. The electronic pulses from the detectors are gated for the period $[t_0, t_0 + T_g]$, with $T_g = 240$ ns, centered on the time determined by the control laser pulse during the retrieval stage. Counts recorded outside the gating period are therefore removed from the analysis. The recorded data allows us to determine the number of photoelectric events $N_2 + N_3$, where N_i is the total number of counts in the i-th channel($i = 1, 2, 3$).

By measuring the retrieved field for different storage times and orientations of the magnetic field, we obtain the collapse and revival signals shown in Fig. 2, (a) through (e). As expected, we observe revivals at integer multiples of the Larmor period. In addition, we see partial revivals at odd multiples of half the Larmor period, except in the vicinity of $\theta = \pi/4$. The measured substructures within a single revival period are in good agreement with the theory (cf., insets of Fig. 2, (e) and (j)). We attribute the overall damping of the revival signal in the experimental data to the magnetic field gradients. The evident decrease of this damping while θ is varied from 0 to $\pi/2$ suggests that such gradients are predominantly along the direction of the signal field wavevector. Similarly, we attribute the additional broadening of the revival peaks at longer times to inhomogeneous magnetic fields, possibly ac fields, not included in the theoretical description. We are pursuing additional investigations to determine the temporal and spatial characteristics of the residual magnetic fields.
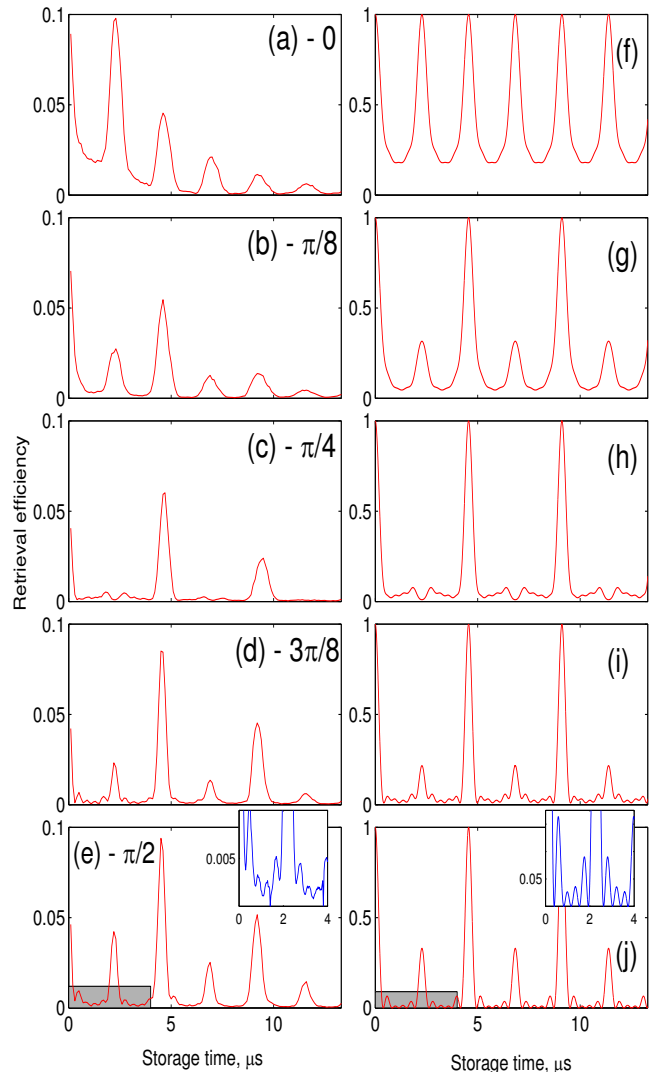

FIG. 2: Panels (a)-(e) show the ratio of the number of photoelectric detection events for the retrieved and incident signal fields for various orientations, $\theta = 0, \pi/8, \pi/4, 3\pi/8, \pi/2$, of the applied magnetic field, and as a function of storage time. The incident signal field is a weak coherent laser pulse. In all cases the control pulse is switched off at $T_s = 0$. We observe a series of collapses and revivals at multiples of the half Larmor period of 2.3 μs. The observed damping over several Larmor periods is likely caused by residual magnetic field gradients. The inset in Panel (e) demonstrates the observed substructure within the first Larmor period. Panels (f) through (j) are corresponding theoretical plots of the dark-state polariton number calculated using Eq.(2).

Theory predicts that both the collapse and the revival times (T_C and T_R, respectively) scale inversely with the magnetic field [11]. In Fig. 3 the theoretical prediction $T_C \approx 0.082T_R$ (solid line) is compared with the experimentally measured values. We find very good agreement except for the lowest value of magnetic field $B = 0.2$ G which may be explained by the presence of residual mag-
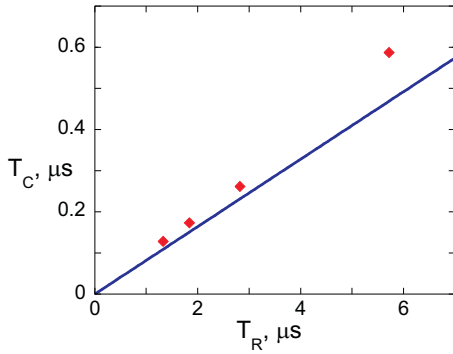
FIG. 3: Diamonds show the measured collapse time T_C of the first revival at half the Larmor period as a function of the measured revival time T_R, for magnetic field values of 0.8, 0.6, 0.4, and 0.2 G, respectively, and for fixed orientation $\theta = \pi/2$. The line shows the corresponding theoretical prediction $T_C \approx 0.082 T_R$ from Eq.(2).
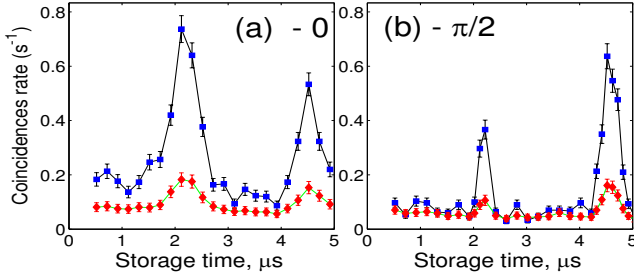
netic field gradients.

FIG. 4: Squares show the measured rate of coincidence detections between D1 and D2,3, $N_{si} = N_{12} + N_{13}$ as a function of the storage time. Diamonds show the measured level of random coincidences N_R. The ratio of squares to diamonds gives g_{si}. Uncertainties are based on the statistics of the photoelectric counting events.

In the measurements presented above, classical, coherent laser light was used as the signal field [29]. We have also investigated the revival dynamics of our atomic memory with the signal field in a single photon state, as shown in Fig. 1. The procedure for production of a single photon state of the signal field at Site A is conditioned on the detection of an idler photon by D1 (see Refs.[10, 26] for further details). If photoelectric detections in different channels $1, 2$ or $1, 3$ happen within the same gating period, they contribute to the corresponding coincidence counts between D1 and Dj, N_{1j}, $j = 2, 3$. We evaluate the α parameter of Grangier et al. [28], given by the ratio of various photoelectric detection probabilities which are measured by the set of detectors D1,D2 and D3. For an ideal single-photon state $\alpha = 0$, whereas for coherent fields $\alpha = 1$. We have experimentally determined $\alpha = 0.51 \pm 0.06$, without any correction for background or dark counts.

The normalized intensity cross-correlation function $g_{si} \equiv (N_{12} + N_{13})/N_R$ may be employed as a measure of non-classical signal-idler field correlations [29, 30], as discussed in detail in Ref.[19]. Here $N_R \equiv N_1 \cdot (N_2 + N_3) \cdot R_{rep}$ is the level of random coincidences, where R_{rep} is the repetition rate of the experimental protocol. The values of g_{si} are obtained by the ratio of the upper and lower traces in Fig. 4. The measurements presented there give values of g_{si} well in excess of two at the revival times, suggesting the dark-state polaritons have a non-classical nature. One could further evaluate self-correlations for the idler field g_{ii}, and for the signal field g_{ss}, and confirm that the Cauchy-Schwarz inequality $g_{si}^2 \leq g_{ss} g_{ii}$ is indeed violated [19, 29, 30]. We have measured, by adding a beamsplitter and an additional detector, the value $g_{ii} = 1.42 \pm 0.03$. When the signal field is stored and retrieved 500 ns later, we find that both $g_{ss} \leq 2$ [10]. While the total number of recorded coincidences between detectors D2 and D3 is not high enough to evaluate g_{ss} for the revived polariton, it is also expected to be less than two, leading to a substantial violation of the Cauchy-Schwarz inequality.

In summary, we have demonstrated revivals of dark-state polaritons in a quantum memory element based on a cold atomic ensemble. The dynamical manipulation and control of collective matter-field excitations, at the level of single quanta, is encouraging for further developments and applications in quantum information science.

[1] H.-J. Briegel, W. Dur, J. I. Cirac, and P. Zoller, Phys. Rev. Lett. **81**, 5932 (1998).
[2] E. Knill, R. Laflamme, and G. J. Milburn, Nature (London) **409**, 46 (2001).
[3] L.-M. Duan, M. D. Lukin, I. J. Cirac, and P. Zoller, Nature **414**, 413 (2001).
[4] A. Kuzmich and E. S. Polzik, in *Quantum Information with Continuous Variables*, eds. S. L. Braunstein and A. K. Pati, Kluwer, (2003).
[5] M. D. Lukin, Rev. Mod. Phys. **75**, 457 (2003).
[6] M. Fleischhauer and M. D. Lukin, Phys. Rev. Lett. **84**, 5094 (2000).
[7] D. F. Phillips et al., Phys. Rev. Lett. **86**, 783 (2001).
[8] C. Liu, Z. Dutton, C. H. Behroozi, L. V. Hau, Nature **409**, 490 (2001).
[9] A. Mair et al., Phys. Rev. A 65, 031802 (2002).
[10] T. Chanelière et al., to be published (2005).
[11] S. D. Jenkins et al., Phys. Rev. A, submitted (2005).
[12] S. E. Harris, Phys. Today **50**, 736 (1997).
[13] M. O. Scully and M. S. Zubairy, *Quantum Optics*, (Cambridge University Press, 1997).

[14] J. Schmiedmayer *et al.*, J. Phys. (France) **4**, 2029 (1994).

[15] G. A. Smith *et al.*, Phys. Rev. Lett. **93**, 163602 (2004).

[16] C. Weisbuch, M. Nishioka, A. Ishikawa, and Y. Arakawa, Phys. Rev. Lett. **69**, 3314 (1992).

[17] J. Jacobson *et al.*, Phys. Rev. A **51**, 2542 (1995).

[18] M. Fleischhauer and M. D. Lukin, Phys. Rev. A **65**, 022314 (2002).

[19] A. Kuzmich *et al.*, Nature, **423**, 726 (2003), and accompanying Supplementary Information.

[20] C. H. van der Wal *et al.*, Science **301**, 196 (2003).

[21] W. Jiang *et al.*, Phys. Rev. A **69**, 043819 (2004).

[22] M. D. Eisaman *et al.*, Phys. Rev. Lett. **93**, 233602 (2004).

[23] C. W. Chou, S. V. Polyakov, A. Kuzmich, and H. J. Kimble, Phys. Rev. Lett. **92**, 213601 (2004).

[24] D. N. Matsukevich and A. Kuzmich, Science **306**, 663 (2004).

[25] V. Balic *et al.*, Phys. Rev. Lett. **94**, 183601 (2005).

[26] D. N. Matsukevich *et al.*, Phys. Rev. Lett. **95**, 040405 (2005).

[27] D. Felinto *et al.*, quant-ph/0507127.

[28] P. Grangier, G. Roger, and A. Aspect, Europhys. Lett. **1**, 173 (1986).

[29] L. Mandel and E. Wolf, *Optical Coherence and Quantum Optics*, (Cambridge University Press, 1995), Chapter 22.

[30] D. F. Walls and G. J. Milburn, *Quantum Optics*, (Springer-Verlag, 1994), Chapters 3 and 5.